

## PATENT COOPERATION TREATY

## PCT

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference <b>1207MD/PCT</b>	<b>FOR FURTHER ACTION</b>	see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.
International application No. <b>PCT/JP 00/02276</b>	International filing date (day/month/year) <b>07/04/2000</b>	(Earliest) Priority Date (day/month/year) <b>09/04/1999</b>
Applicant <b>DU PONT-MITSUI POLYCHEMICALS CO., LTD. et al.</b>		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 3 sheets.

It is also accompanied by a copy of each prior art document cited in this report.

**1. Basis of the report**

- a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.
  - the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).
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  - the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
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2.  **Certain claims were found unsearchable (See Box I).**

3.  **Unity of Invention is lacking (see Box II).**

4. With regard to the **title**,

- the text is approved as submitted by the applicant.
- the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,

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6. The figure of the **drawings** to be published with the abstract is Figure No.

- as suggested by the applicant.
- because the applicant failed to suggest a figure.
- because this figure better characterizes the invention.

None of the figures.

## INTERNATIONAL SEARCH REPORT

National Application No

PCT/JP 00/02276

**A. CLASSIFICATION OF SUBJECT MATTER**  
 IPC 7 C08J5/18 B65D65/02 //C08L23:08

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08J B65D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

WPI Data, PAJ, EPO-Internal

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 91 16376 A (DU PONT) 31 October 1991 (1991-10-31) claims 1,3,4,6-8 examples ---	1-4,7-9
A	DATABASE WPI Section Ch, Week 199506 Derwent Publications Ltd., London, GB; Class A17, AN 1995-041430 XP002141885 & JP 06 322192 A (SHINETSU POLYMER KK), 22 November 1994 (1994-11-22) abstract ---	1
A	EP 0 521 426 A (BASF AG) 7 January 1993 (1993-01-07) claims 1,3,4 ---	1 -/-

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

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Date of the actual completion of the international search Date of mailing of the international search report

5 July 2000

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## INTERNATIONAL SEARCH REPORT

International Application No  
PCT/JP 00/02276

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DATABASE WPI Section Ch, Week 199610 Derwent Publications Ltd., London, GB; Class A18, AN 1996-094359 XP002141886 & JP 08 003386 A (MITSUI DU PONT POLYCHEMICAL KK), 9 January 1996 (1996-01-09) abstract --- EP 0 664 317 A (DU PONT INT) 26 July 1995 (1995-07-26) claims 1,2,11 page 2, line 38 - line 45 -----	1
A		1

**INTERNATIONAL SEARCH REPORT**

Information on patent family members

International Application No

PCT/JP 00/02276

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
WO 9116376	A 31-10-1991	CS 9101167 A		15-01-1992
		EP 0483151 A		06-05-1992
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EP 0521426	A 07-01-1993	DE 4122313 A		14-01-1993
		DE 59205440 D		04-04-1996
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		EP 0740684 A		06-11-1996
		JP 9508171 T		19-08-1997
		WO 9520624 A		03-08-1995
		US 5750611 A		12-05-1998

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## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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(54) Title: FILM FOR WRAPPING

(57) Abstract

A film having a large tensile stress in the machine direction, excellent mechanical properties and stretch-wrapping property, and that is less broken at the time of wrapping, can be favorably torn off in the transverse direction, can be intimately adhered on the container walls to be wrapped, and excellently restores its shape after it is depressed and deformed as a result of wrapping. The film for stretch-wrapping is formed of a resin composition containing, as a chief component, an ethylene/(meth)acrylic acid/(meth)acrylic acid ester terpolymer that contains not more than 7% by weight of a (meth)acrylic acid ester unit, and, optionally, containing an anti-fogging agent or a tackifier.

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## FILM FOR WRAPPING

### (Technical Field)

This application claims priority to Japanese Patent  
5 Application No. 11-103239 filed on April 9, 1999, which is  
incorporated herein by reference.

The present invention relates to a film for stretch-wrapping. More particularly, the invention relates to a film for stretch-wrapping comprising chiefly an ethylene -  
10 (meth)acrylic acid - (meth)acrylic acid ester terpolymer or a metal salt ionomer thereof.

### (Background Art)

PVC films have heretofore been chiefly used as stretchable films (for wrapping foamed polystyrene trays  
15 containing foods) for business use such as in supermarkets and convenience stores. However, from the environmental viewpoints in recent years, it has been urged to provide a substitute for the PVC. In the field of stretchable films for stretch-wrapping, the PVC films have rapidly been  
20 substituted by the olefin resin films.

Many films for stretch-wrapping have so far been proposed using an ethylene/unsaturated carboxylic acid copolymer, an ethylene/unsaturated carboxylic acid/unsaturated carboxylic acid ester terpolymer or an  
25 ionomer thereof.

Japanese Unexamined Patent Publication (Kokai) No. 134591/1978 discloses a film for wrapping of an ethylene copolymer comprising (a) an ethylene, (b) an unsaturated carboxylic acid alkyl ester, (c) an unsaturated carboxylic acid and (d) a metal salt of an unsaturated carboxylic acid, the component (a) being contained in an amount of from 90 to 98 mol%, the component (b) being contained in an amount of from 9.7 to 2.0 mol%, the component (c) being contained in an amount of from 0 to 2.5 mol%, and the  
30 component (d) being contained in an amount of from 0.3 to  
35

2.5 mol%.

Japanese Laid-Open Patent Publication for PCT application (Kohyo) No. 506820/1992 discloses a film for wrapping comprising:

- 5       (a) at least 80% of a terpolymer of at least 50% by weight of an ethylene, 2 to 20% by weight of an unsaturated carboxylic acid having 3 to 8 carbon atoms, and 2 to 20% by weight of at least one kind of an alkyl acrylate or an alkyl methacrylate in which the alkyl group  
10 has 2 to 12 carbon atoms, or a moiety derived from a mixture thereof, 0 to 10% of the acid group of the acid group-containing moiety thereof being neutralized with at least one kind of metal ions; and  
15       (b) 0.1 to 2% by weight of at least one kind of a sorbitan fatty acid ester;

at least one surface of said film being treated with corona to a degree sufficient for producing a wet tensile force of 40 to 50 dynes/cm on the treated surface thereof.

- Japanese Examined Patent Publication (Kokoku) No.
- 20 100741/1995 discloses a film for stretch-wrapping obtained by inflation-molding an ethylene methacrylate copolymer resin containing 8 to 12% by weight of a methacrylic acid and having a tensile stress of 1.1 to 1.5 times in both the machine and transverse directions when stretched by  
25 100% and having a tensile stress of 1.5 to 2.0 times in the transverse direction when stretched by 200% with respect to the stress of when it is stretched by 50%.

- Japanese Patent No. 2642583 discloses a film for stretch-wrapping comprising chiefly 50 to 99% by weight of an ethylene - (meth)acrylic acid copolymer resin and 50 to 1% by weight of an ethylene - (meth)acrylic acid - (meth)acrylic acid ester terpolymer resin and, optionally, containing an anti-fogging agent (a defogging agent) or a tackifier. There have been described that the above  
35 bipolymer alone or the terpolymer alone exhibits a 100%

tensile stress that does not lie within a suitable range (110 to 170 kg/cm<sup>2</sup> in the machine direction of the film, and 60 to 110 kg/cm<sup>2</sup> in the transverse direction), and is not suited for the automatic wrapping.

5       The ethylene - (meth)acrylic acid bipolymer generally has a high stiffness, whereas the ethylene - (meth)acrylic acid - (meth)acrylic acid ester terpolymer has a low stiffness and is soft. This holds even for the ionomers obtained by neutralizing the above copolymers with metal 10 ions. The ionomer of the above bipolymer is called hard ionomer, and the ionomer of the above terpolymer is called soft ionomer.

15      The film for stretch-wrapping disclosed in the specification of Japanese Patent No. 2642583 is obtained by mixing the above bipolymer and the terpolymer at a predetermined ratio so as to be suited for the automatic wrapping. However, these two copolymers are not necessarily compatible to each other and, hence, the external haze value tends to increase. Therefore, the 20 film is not still satisfactory from the standpoint of seeing through the wrapped content.

25      Further, the film for stretch-wrapping is usually blended with an anti-fogging agent to improve the property for seeing through the content. When the ionomers of the above-mentioned copolymers are used, however, gel tends to be formed during the molding operation with the passage of time, making it difficult to prepare the film itself.

(Summary of the Invention)

30      It is therefore an object of the present invention to provide a film constituted by the above-mentioned terpolymer or an ionomer thereof, having a large tensile stress in the machine direction, excellent mechanical properties and stretch-wrapping property, i.e., to provide a film for stretch-wrapping that is less broken at the 35 time of wrapping, that can be favorably torn off in the

transverse direction, that can be intimately adhered on the container walls to be wrapped, and that excellently restores its shape after it is depressed and deformed as a result of wrapping.

5 Another object of the present invention is to provide a film for stretch-wrapping that can be easily formed and easily treated, that is homogeneous even optically, exhibiting less external haze and enabling the content to be seen through.

10 A further object of the present invention is to provide a film for stretch-wrapping, which suppresses the gelling tendency when it is blended with an anti-fogging agent without impairing the appearance or without being foamed during the working.

15 According to the present invention, there is provided a film for stretch-wrapping formed of a resin composition containing, as a chief component, an ethylene/(meth)acrylic acid/(meth)acrylic acid ester terpolymer that contains not more than 7% by weight and, 20 particularly, less than 5% by weight of a (meth)acrylic acid ester unit, and, optionally, containing an anti-fogging agent or a tackifier.

It is desired that the terpolymer used in the present invention contains from 5 to 20% by weight of a 25 (meth)acrylic acid, and not less than 0.1% by weight but less than 5% by weight of a (meth)acrylic acid ester.

The film for stretch-wrapping can be prepared by the inflation method. However, the terpolymer used in the invention can be molded into a film by the T-die method 30 offering an advantage that the film can be produced at a high speed.

According to the present invention, there is further provided a film for stretch-wrapping formed of a resin composition containing, as a chief component, an ionomer obtained by ionizing an ethylene/(meth)acrylic 35

acid/(meth)acrylic acid ester terpolymer that contains less than 5% by weight of a (meth)acrylic acid ester unit, and, optionally, containing an anti-fogging agent or a tackifier.

5 It is desired that the ionomer used in the invention uses, as a base, a copolymer that contains from 5 to 20% by weight of a (meth)acrylic acid and not less than 0.1% by weight but less than 5% by weight of a (meth)acrylic acid ester, and has an ionization degree of from 0.1 to  
10 30%.

This ionomer, too, has an advantage that it can be molded into a film by the T-die method.

(Detailed Description of the Invention)

[Action]

15 The present invention uses an ethylene/(meth)acrylic acid/(meth)acrylic acid ester terpolymer or an alkali metal ionomer thereof as a resin for the film for stretch-wrapping, containing a (meth)acrylic acid ester in a limited amount such as not larger than 7% by weight and,  
20 particularly, smaller than 5% by weight.

As will be described later in Examples appearing later, it was learned that the film for stretch-wrapping formed of the terpolymer or the ionomer thereof exhibits a stress of generally from 20 to 40 MPa (196 to 392 kg/cm<sup>2</sup>)  
25 when stretched by 100% in the machine direction, which is a value considerably higher than that of the traditionally used films for stretch-wrapping, thus exhibiting excellent mechanical properties and adaptability to stretch-wrapping.

30 That is, the film for stretch-wrapping exhibits not only improved mechanical strength in the machine direction and excellent moldability but also distinguished advantage of decreasing the thickness of the film, decreased film breakage at the time of wrapping, excellent tear-off  
35 property in the transverse direction of the film,

excellent adhesiveness to the container walls that are to be wrapped, and excellent restoration after depressed and deformed as a result of wrapping.

The resin for film used in the invention is prepared  
5 by the direct polymerization of an ethylene, a (meth)acrylic acid and a (meth)acrylic acid ester, and has very homogeneous composition and distribution as compared with a resin composition prepared by blending a bipolymer and a terpolymer, can be favorably molded into the film  
10 and can be favorably worked. Besides, the film for stretch-wrapping that is formed is homogeneous even optically, exhibiting less external haze and offering advantage that the content can be favorably seen through.

The present invention uses the ionomer of the above  
15 terpolymer. Here, however, it is important that the ionomer is the one that is neutralized with an alkali metal. The ionomer has a structure in which the copolymer is ionically crosslinked at the portions of acid groups, and it is believed that the ionically crosslinked  
20 structure helps improve the above-mentioned adaptability to stretch-wrapping without spoiling the moldability into films. As a cationic seed used for introducing the ionically crosslinked structure, there can be used an alkali metal, an alkaline earth metal or zinc. In the  
25 ionomer using zinc or alkaline earth metal, however, there takes place a reaction with the anti-fogging agent blended in the film, deteriorating the film-forming property and appearance. According to the present invention, on the other hand, an alkali metal is selected as a metal seed  
30 for the ionomer, suppressing the reaction with the anti-fogging agent and improving the film-forming property and appearance.

[Terpolymer]

The present invention uses an ethylene/(meth)acrylic acid/(meth)acrylic acid ester terpolymer containing not  
35

more than 7% by weight and, most desirably, from 0.1 to (less than) 5% by weight of a (meth)acrylic acid ester.

When the bipolymer without containing the (meth)acrylic acid ester unit is used, the film exhibits 5 increased stiffness and is broken at the time of wrapping and is restored little after the film is depressed and deformed as a result of wrapping.

When the content of the (meth)acrylic acid ester exceeds the above range, on the other hand, the object of 10 the present invention is not accomplished which is to provide a high-stress film for stretch-wrapping, and the adaptability to stretch-wrapping decreases.

In the present invention, examples of the (meth)acrylic acid ester used for the terpolymer include 15 methyl acrylate, ethyl acrylate, isopropyl acrylate, n-butyl acrylate, isobutyl acrylate, isoctyl acrylate, methyl methacrylate, ethyl methacrylate and isobutyl methacrylate.

Among these (meth)acrylic acid esters, there can be 20 preferably used a (meth)acrylic acid ester in which the alkyl group has 1 to 10 carbon atoms and, particularly, an isobutyl (meth)acrylate.

It is desired that the terpolymer contains the (meth)acrylic acid in an amount of from 5 to 20% by weight 25 and, preferably, from 8 to 15% by weight.

When the content of the (meth)acrylic acid is smaller than the above range, the stress when the film is stretched tends to become smaller than the above range. When the content of the (meth)acrylic acid is larger than 30 the above range, on the other hand, the stress when the film is stretched tends to exceed the above range. In either case, the adaptability to stretch-wrapping becomes inferior as compared to when the content lies within the above-mentioned range.

35 It is desired that the terpolymer has a melt flow

rate (JIS K6760) of from 0.1 to 100 g/10 minutes and, particularly, from 0.2 to 30 g/10 minutes from the standpoint of mechanical properties of the film and moldability into films.

5       The terpolymer used in the present invention is prepared by the direct polymerization of an ethylene, a (meth)acrylic acid and a (meth)acrylic acid ester like the method of preparing a high-pressure method polyethylene.  
[Ionomer]

10      In the present invention, the alkali metal ionomer of the terpolymer, too, is used as the resin for forming the film. The terpolymer has the composition and properties as described above.

15      The ethylene/(meth)acrylic acid/(meth)acrylic acid ester terpolymer has an ionization degree of from 0.1 to 30%, preferably, from 0.1 to 10% and, particularly preferably, from 0.1 to 5%.

20      When the ionization degree exceeds the above range, the ionomer absorbs moisture developing inconvenience such as foaming at the time of machining.

The importance for using the alkali metal as an ionic seed for the ionomer was described already. As the alkali metal in the ionomer, there can be exemplified lithium, sodium and potassium.

25      The terpolymer can be ionized by using various kinds of compounds of an alkali metal, such as oxide, hydroxide, carbonate, bicarbonate, salt of fatty acid and the like. The ionization is conducted in accordance with a widely known method of preparing ionomers.

30      It is desired that the ionomer has a melt flow rate (JIS K6760) of from 0.1 to 100 g/10 min. and, particularly, from 0.2 to 30 g/10 min. from the standpoint of mechanical properties and moldability into films.

[Film for stretch-wrapping and method of its preparation]

35      The film for stretch-wrapping can be prepared by

melt-extruding the above-mentioned terpolymer or the ionomer thereof and molding it into a film.

The resin for forming the film comprises, as a chief component, the above-mentioned terpolymer or the ionomer thereof. Concretely, the film may comprise greater than 5 50 wt%, preferably greater than 70 wt% of the terpolymer or the ionomer thereof. Most preferably, the resin for forming the film consists essentially of the terpolymer or the ionomer thereof.

10 The resin for forming the film may be optionally blended with an anti-fogging agent for better see through of the content irrespective of the presence of water or a tackifier for imparting tackiness to the film.

15 As the anti-fogging agent, there can be exemplified any anti-fogging agent that has been known per se. such as the one of the type of glycerin fatty acid ester, sorbitan fatty acid ester, di- or polyglycerin fatty acid ester and ethylene oxide adduct, to which only, however, the anti-fogging agent is in no way limited.

20 The anti-fogging agent can be blended in an amount of from 0.1 to 10 parts by weight and, particularly, from 0.1 to 8 parts by weight per 100 parts by weight of the resin for forming the film.

25 As the tackifier, there can be exemplified aliphatic hydrocarbon resin, aromatic hydrocarbon resin, aliphatic/aromatic copolymerizable hydrocarbon resin, alicyclic hydrocarbon resin, synthetic terpene hydrocarbon resin, terpene hydrocarbon resin, cumarone-indene hydrocarbon resin, low-molecular styrene resin, rosin 30 hydrocarbon resin, or combinations thereof, to which only, however, the tackifier is not limited, as a matter of course.

35 The tackifier can be blended in an amount of from 0.1 to 15 parts by weight and, particularly, from 0.1 to 10 parts by weight per 100 parts by weight of the resin for

forming the film.

The resin for forming the film of the present invention can be blended with widely known resin blending agents, such as coloring agent, heat stabilizer, weather 5 resistance stabilizer, antioxidant, anti-aging agent, photo-stabilizer, ultraviolet absorber, anti-static agent, lubricant such as metal soap or wax, reforming resin or rubber, and the like according to a known recipe.

The film for stretch-wrapping of the present 10 invention has a feature in that it exhibits a stress of from 20 to 40 MPa when it is stretched by 100% in the machine direction (MD) thus exhibiting a stress that lies within a high range when it is stretched in the mechanical direction.

15 The film for stretch-wrapping of the present invention, on the other hand, exhibits a stress that is suppressed to lie within a low range of from 5 to 20 MPz when it is stretched by 100% in the transverse direction (TD).

20 Thus, the film for stretch-wrapping of the present invention exhibits a ratio (MD/TD) of the stress in the machine direction to the stress in the transverse direction of from 2 to 8 when it is stretched by 100%, featuring a large anisotropy in the tensile stress and, 25 hence, offering advantage as described above and advantage in the productivity as will be described later.

The film for stretch-wrapping of the invention 30 exhibits a large tensile stress in the machine direction and permits the thickness of the film to be considerably decreased. The film can be used having a thickness of, generally, from 5 to 20  $\mu\text{m}$  and, particularly, from 7 to 18  $\mu\text{m}$  though it may vary depending upon the applications.

The film for stretch-wrapping of the invention can be produced by the inflation method. However, the terpolymer 35 used in the invention can be formed into a film by the T-

die method at a high speed, which is an advantage.

In forming the film, the resin is heated at a temperature higher than its melting point but lower than its decomposition temperature, and is, generally, heated 5 over a range of from 180 to 240°C.

That is, the terpolymer or the ionomer thereof is formed into the film by the T-die cast method, and the film that is formed is taken up at a high speed so as to be stretched in the machine direction. According to the 10 present invention, therefore, the film can be produced at a speed of 150 m/min or faster, which is superior in the productivity to the production at a speed of 50 m/min. at the greatest by the inflation method.

The terpolymer and the ionomer thereof used in the 15 present invention exhibits excellent moldability, and give no inconvenience when it is molded into a film for stretch-wrapping even by the inflation method.

(EXAMPLES)

The present invention will now be concretely 20 described by way of Examples to which only, however, the invention is in no way limited.

1. Starting materials.

(1) Ethylene/methacrylic acid/isobutyl acrylate copolymer (E/MAA/iBA).

25	Methacrylic acid	10% by weight
	Isobutyl acrylate	3% by weight
	MFR	8.5 g/10 min.

(2) Ionomer ①

Base polymer: ethylene/methacrylic acid/isobutyl acrylate 30 copolymer (methacrylic acid = 10% by weight, isobutyl acrylate = 3% by weight)

Metallic ion seed	sodium
Ionization degree	2%
MFR	7.9 g/10 min.

35 (3) Ionomer ②

Base polymer: ethylene/methacrylic acid/isobutyl acrylate copolymer (methacrylic acid = 10% by weight, isobutyl acrylate = 10% by weight)

	Metallic ion seed	sodium
5	Ionization degree	36%
	MFR	1.1 g/10 min.

(4) Ionomer ③

Base polymer: ethylene/methacrylic acid copolymer (methacrylic acid = 10% by weight)

10	Metallic ion seed	sodium
	Ionization degree	50%
	MFR	1.3 g/10 min.

(5) Ionomer ④

Base polymer: ethylene/methacrylic acid/isobutyl acrylate copolymer (methacrylic acid = 10% by weight, isobutyl acrylate = 3% by weight)

	Metallic ion seed	magnesium
	Ionization degree	2%
	MFR	8.0 g/10 min.

20 (6) Ionomer ⑤

Base polymer: ethylene/methacrylic acid/isobutyl acrylate copolymer (methacrylic acid = 10% by weight, isobutyl acrylate = 3% by weight)

	Metallic ion seed	zinc
25	Ionization degree	2%
	MFR	8.0 g/10 min.

2. Working method.

A mixture of 98% by weight of the above resin and 2% by weight of a diglycerin oleate (O-71DE manufactured by Riken Vitamin Co.) was formed into films using the apparatuses described below.

\* T-die method.

	Extruder	65 mm in diameter (L/D = 32)
		screw full-flighted type
35	Die	coat hanger type (900 mm wide)

The working temperature was set to 225°C (resin temperature) and the thickness of the film was selected to be 12 µm. Further, the take-up speed was set to be 165 m/min.

5 \*Inflation method.

Extruder                50 mm in diameter (L/D = 28)  
                          screw     3-stage type

Die                      150 mm in diameter, spiral type

The working temperature was set to 200°C (resin temperature) and the thickness of the film was selected to be 12 µm. Further, the take-up speed was set to be 20 m/min.

3. Items to be Evaluated and Results.

Moldability during the working was evaluated concerning the following items. The results were as shown in Tables 1 and 2.

(1) Reactivity with the anti-fogging agent.

It was examined whether a gel was formed upon the reaction with the anti-fogging agent during the molding.

20                      ○: No gel was formed  
                          X: Gel was formed with the passage of time

(2) Cleavage in the film.

It was examined whether the film was cleaved due to scars in the film during the molding.

25                      ○: Could be stably worked at a high speed.  
                          X: Cleaved along the scars during the working, and could not be stably worked at a high speed.

(3) Appearance of the film.

30                      The appearance of the film was examined during the molding.

○: The film was maintained smooth.  
X: The surface of the film became rugged due to gel and lumps.

35                      The handling of the pellets used was evaluated

concerning the following items. The results were as shown in Tables 1 and 2.

(1) Hygroscopic property.

In order to evaluate defects such as foaming, etc. at 5 the time of forming the ionomer into a film, the saturated amount of water was measured when the pellets were left to stand on the site of working.

10           ○: The saturated amount of water was smaller than 1000 ppm in an atmosphere of 30°C, 90%RH.

              ×: The saturated amount of water exceeded 1000 ppm under the same conditions.

The films that were formed were measured for their film properties under the following conditions.

15 (1) Stress of when the film was stretched.

Dumbbell   No.1 dumbbell specified under JIS K7113  
Crosshead speed 200 m/min.

20 The wrapping test was conducted by using trays of a foamed styrene and an automatic wrapping machine (AW-3600 manufactured by Teraoka Seikosha Co.) to evaluate the following items. The results were as shown in Tables 1 and 2.

(1) Film cutting property.

25 The film was cut using a paper saw or a metal saw to evaluate the easiness of cutting.

○: The film was sharply cut in the transverse direction.

×: The film was not sharply cut in the transverse direction.

30 (2) Breakage of film.

The wrapping test was conducted while variously changing the tension of the film to observe the appearance.

○: The film was not broken.

35           ×: The film was broken.

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## (3) Wrinkles on the film when packaged.

Adhesion to the foamed styrene container was observed.

○: Film adhered tightly without floating.

5           ×: Film floated.

## (4) Restoring property after depressed with fingers.

The upper surface of the film wrapping the container was depressed with a predetermined pressure by a finger.

The finger was then released so that the film restored,

10 and the mark made by the finger was observed.

○: There remained no mark of finger.

×: There remained the mark of finger.

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Table 1

5	<u>Example 1</u>	<u>Example 2</u>	<u>Example 3</u>	<u>Example 4</u>
Resin	E/MAA/iBA	E/MAA/iBA	ionomer ①	ionomer ②
Working method	T-die	inflation	T-die	inflation
Reactivity				
10 with anti-				
fogging agent	○	○	○	○
Cutting				
property of				
film	○	○	○	○
15 Appearance				
of film	○	○	○	○
Hygroscopic				
property of				
pellets	○	○	○	○
20 Film properties				
Tensile stress				
50%	20/6.6	17/7.0	21/6.7	18/7.1
(MD/TD) 100%	25/7.2	22/7.5	26/7.5	23/7.7
[Mpa] 200%	--/8.8	--/8.8	--/8.9	--/8.6
25 Adaptability				
to wrapping				
machine Cutting				
property	○	○	○	○
Breakage	○	○	○	○
30 Wrinkles	○	○	○	○
Restoration				
after depressed				
with finger	○	○	○	○

Table 2

	<u>Comp.Ex.1</u>	<u>Comp.Ex.2</u>	<u>Comp.Ex.3</u>	<u>Comp.Ex.4</u>
5 Resin	ionomer ②	ionomer ③	ionomer ④	ionomer ⑤
Working method	T-die	T-die	T-die	T-die
Reactivity				
10 with anti-				
fogging agent	○	○	x	x
Cutting				
property of				
film	○	○	x	x
15 Appearance of				
film	○	○	x	x
Hygroscopic				
property of				
pellets	x	x	○	○
20 Tensile stress				
50%	--/--	--/--	--/--	--/--
(MD/TD)	--/--	--/--	--/--	--/--
[MPa]	--/--	--/--	--/--	--/--
Adaptability				
25 to wrapping				
machine Cutting				
property	--	--	--	--
Breakage	--	--	--	--
Wrinkles	--	--	--	--
30 Restoration				
after depressed				
with finger	--	--	--	--

## (Field of Utilization in Industry)

According to the present invention, an ethylene/(meth)acrylic acid/(meth)acrylic acid ester terpolymer containing a (meth)acrylic acid ester in an amount within a predetermined range or an ionomer thereof is used for a film for stretch-wrapping. Therefore, the film for stretch-wrapping exhibits a large tensile stress in the machine direction, excellent mechanical properties and excellent adaptability to stretch-wrapping.

That is, the film breaks little during the wrapping, can be favorably torn off in the transverse direction, intimately and favorably adheres onto the walls of the container that is to be wrapped, and favorably restores after depressed and deformed as a result of wrapping.

The resin for forming the film is produced by the direct polymerization of the above-mentioned three components, has a homogeneous composition, can be favorably molded into a film and can be favorably worked. Besides, the film for stretch-wrapping that is formed is homogeneous even optically, exhibits less external haze and excellently permits the content to be seen through.

Even when the ionomer is used as a resin for forming the film, the gelling tendency is suppressed when the anti-fogging agent is blended, exhibiting excellent appearance and hygroscopic property.

Thus, the film for stretch-wrapping of the invention is useful for wrapping sea foods, meats, vegetables, fruits and daily dishes placed on or contained in the plastic trays.

## Claims

1. A film for stretch-wrapping formed of a resin composition containing, as a chief component, an ethylene/(meth)acrylic acid/(meth)acrylic acid ester terpolymer that contains not more than 7% by weight of a (meth)acrylic acid ester unit, and, optionally, containing an anti-fogging agent or a tackifier.
- 5 2. A film for stretch-wrapping formed of a resin composition containing, as a chief component, an ethylene/(meth)acrylic acid/(meth)acrylic acid ester terpolymer that contains less than 5% by weight of a (meth)acrylic acid ester unit, and, optionally, containing an anti-fogging agent or a tackifier.
- 10 3. A film for stretch-wrapping according to claim 1 or 2, wherein said terpolymer is the one that contains from 5 to 20% by weight of a (meth)acrylic acid, and not less than 0.1% by weight but less than 5% by weight of a (meth)acrylic acid ester.
- 15 4. A film for stretch-wrapping according to any one of claims 1 to 3, wherein the alkyl group of the (meth)acrylic acid ester has from 1 to 10 carbon atoms.
- 20 5. A film for stretch-wrapping according to any one of claims 1 to 4, wherein the stress in the machine direction of when the film is stretched by 100% lies within a range of from 20 to 40 MPa.
- 25 6. A film for stretch-wrapping according to any one of claims 1 to 6, wherein the molding is effected according to the T-die method.
- 30 7. A film for stretch-wrapping formed of a resin composition containing, as a chief component, an ionomer obtained by ionizing with an alkali metal, an ethylene/(meth)acrylic acid/(meth)acrylic acid ester terpolymer that contains less than 5% by weight of a (meth)acrylic acid ester unit, and, optionally, containing
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an anti-fogging agent or a tackifier.

8. A film for stretch-wrapping according to claim 7, wherein said terpolymer is the one that contains from 5 to 20% by weight of a (meth)acrylic acid, and not less than 5 0.1% by weight but less than 5% by weight of a (meth)acrylic acid ester, and the ionomer has an ionization degree of from 0.1 to 30%.

9. A film for stretch-wrapping according to claim 6 or 7, wherein the alkyl group of the (meth)acrylic acid 10 ester has from 1 to 10 carbon atoms.

10. A film for stretch-wrapping according to any one of claims 7 to 9, wherein the stress in the machine direction of when the film is stretched by 100% lies within a range of from 20 to 40 MPa.

15 11. A film for stretch-wrapping according to any one of claims 7 to 10, wherein the forming of the film is effected according to the T-die method.

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## INTERNATIONAL SEARCH REPORT

International Application No

PCT/JP 00/02276

A. CLASSIFICATION OF SUBJECT MATTER  
 IPC 7 C08J/18 B65D65/02 //C08L23:08

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
 IPC 7 C08J B65D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

WPI Data, PAJ, EP0-Internal

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 91 16376 A (DU PONT) 31 October 1991 (1991-10-31) claims 1,3,4,6-8 examples ----	1-4,7-9
A	DATABASE WPI Section Ch, Week 199506 Derwent Publications Ltd., London, GB; Class A17, AN 1995-041430 XP002141885 & JP 06 322192 A (SHINETSU POLYMER KK), 22 November 1994 (1994-11-22) abstract ----	1
A	EP 0 521 426 A (BASF AG) 7 January 1993 (1993-01-07) claims 1,3,4 ----	1 -/-

 Further documents are listed in the continuation of box C. Patent family members are listed in annex.

## \* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search

Date of mailing of the international search report

5 July 2000

18/07/2000

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## INTERNATIONAL SEARCH REPORT

International Application No

PCT/JP 00/02276

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>DATABASE WPI            Section Ch, Week 199610            Derwent Publications Ltd., London, GB;            Class A18, AN 1996-094359            XP002141886            &amp; JP 08 003386 A (MITSUI DU PONT            POLYCHEMICAL KK),            9 January 1996 (1996-01-09)            abstract</p> <p>-----</p>	1
A	<p>EP 0 664 317 A (DU PONT INT)            26 July 1995 (1995-07-26)            claims 1,2,11            page 2, line 38 - line 45</p> <p>-----</p>	1

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/JP 00/02276

Patent document cited in search report	Publication date		Patent family member(s)		Publication date
WO 9116376	A	31-10-1991	CS EP HU JP PL	9101167 A 0483151 A 62625 A 4506820 T 290013 A	15-01-1992 06-05-1992 28-05-1993 26-11-1992 02-12-1991
JP 6322192	A	22-11-1994	JP	2642583 B	20-08-1997
EP 0521426	A	07-01-1993	DE DE	4122313 A 59205440 D	14-01-1993 04-04-1996
JP 8003386	A	09-01-1996	NONE		
EP 0664317	A	26-07-1995	DE EP JP WO US	69512382 D 0740684 A 9508171 T 9520624 A 5750611 A	28-10-1999 06-11-1996 19-08-1997 03-08-1995 12-05-1998

**VERTRAG ÜBER DIE INTERNATIONALE ZUSAMMENARBEIT  
AUF DEM GEBIET DES PATENTWESENS**

**PCT**

**INTERNATIONALER RECHERCHENBERICHT**

(Artikel 18 sowie Regeln 43 und 44 PCT)

Aktenzeichen des Anmelders oder Anwalts <b>M/Z+J-133-PC</b>	<b>WEITERES VORGEHEN</b>	siehe Mitteilung über die Übermittlung des internationalen Recherchenberichts (Formblatt PCT/ISA/220) sowie, soweit zutreffend, nachstehender Punkt 5
Internationales Aktenzeichen <b>PCT/EP 00/ 02821</b>	Internationales Anmelde datum (Tag/Monat/Jahr) <b>30/03/2000</b>	(Frühestes) Prioritätsdatum (Tag/Monat/Jahr) <b>08/04/1999</b>
Anmelder		
<b>ZIMMERMANN &amp; JANSEN GMBH</b>		

Dieser internationale Recherchenbericht wurde von der Internationalen Recherchenbehörde erstellt und wird dem Anmelder gemäß Artikel 18 übermittelt. Eine Kopie wird dem Internationalen Büro übermittelt.

Dieser internationale Recherchenbericht umfaßt insgesamt 2 Blätter.

Darüber hinaus liegt ihm jeweils eine Kopie der in diesem Bericht genannten Unterlagen zum Stand der Technik bei.

**1. Grundlage des Berichts**

- a. Hinsichtlich der **Sprache** ist die internationale Recherche auf der Grundlage der internationalen Anmeldung in der Sprache durchgeführt worden, in der sie eingereicht wurde, sofern unter diesem Punkt nichts anderes angegeben ist.
  - Die internationale Recherche ist auf der Grundlage einer bei der Behörde eingereichten Übersetzung der internationalen Anmeldung (Regel 23.1 b)) durchgeführt worden.
- b. Hinsichtlich der in der internationalen Anmeldung offenbarten **Nucleotid- und/oder Aminosäuresequenz** ist die internationale Recherche auf der Grundlage des Sequenzprotokolls durchgeführt worden, das
  - in der internationalen Anmeldung in Schriftlicher Form enthalten ist.
  - zusammen mit der internationalen Anmeldung in computerlesbarer Form eingereicht worden ist.
  - bei der Behörde nachträglich in schriftlicher Form eingereicht worden ist.
  - bei der Behörde nachträglich in computerlesbarer Form eingereicht worden ist.
  - Die Erklärung, daß das nachträglich eingereichte schriftliche Sequenzprotokoll nicht über den Offenbarungsgehalt der internationalen Anmeldung im Anmeldezeitpunkt hinausgeht, wurde vorgelegt.
  - Die Erklärung, daß die in computerlesbarer Form erfaßten Informationen dem schriftlichen Sequenzprotokoll entsprechen, wurde vorgelegt.

2.  **Bestimmte Ansprüche haben sich als nicht recherchierbar erwiesen** (siehe Feld I).

3.  **Mangelnde Einheitlichkeit der Erfindung** (siehe Feld II).

**4. Hinsichtlich der Bezeichnung der Erfindung**

- wird der vom Anmelder eingereichte Wortlaut genehmigt.
- wurde der Wortlaut von der Behörde wie folgt festgesetzt:

**5. Hinsichtlich der Zusammenfassung**

- wird der vom Anmelder eingereichte Wortlaut genehmigt.
- wurde der Wortlaut nach Regel 38.2b) in der in Feld III angegebenen Fassung von der Behörde festgesetzt. Der Anmelder kann der Behörde innerhalb eines Monats nach dem Datum der Absendung dieses internationalen Recherchenberichts eine Stellungnahme vorlegen.

**6. Folgende Abbildung der Zeichnungen ist mit der Zusammenfassung zu veröffentlichen: Abb. Nr. 3a**

- wie vom Anmelder vorgeschlagen
- weil der Anmelder selbst keine Abbildung vorgeschlagen hat.
- weil diese Abbildung die Erfindung besser kennzeichnet.

 keine der Abb.

# INTERNATIONALER RECHERCHENBERICHT

Internationales Aktenzeichen

PCT/EP 00/02821

## A. KLASSEFIZIERUNG DES ANMELDUNGSGEGENSTANDES

IPK 7 C10G11/18 F16K1/16 F16K1/22 B65D90/62

Nach der Internationalen Patentklassifikation (IPK) oder nach der nationalen Klassifikation und der IPK

## B. RECHERCHIERTE GEBIETE

Recherchierte Mindestprässtoff (Klassifikationssystem und Klassifikationssymbole)

IPK 7 C10G F16K B65D

Recherchierte aber nicht zum Mindestprässtoff gehörende Veröffentlichungen, soweit diese unter die recherchierten Gebiete fallen

Während der internationalen Recherche konsultierte elektronische Datenbank (Name der Datenbank und evtl. verwendete Suchbegriffe)

EPO-Internal, WPI Data, PAJ

## C. ALS WESENTLICH ANGESEHENE UNTERLAGEN

Kategorie°	Bezeichnung der Veröffentlichung, soweit erforderlich unter Angabe der in Betracht kommenden Teile	Betr. Anspruch Nr.
X	US 4 489 862 A (DIEM WINFRIED) 25. Dezember 1984 (1984-12-25)	7,8
Y	Ansprüche 1,2,5 Abbildung 3 Spalte 6, Zeile 57 -Spalte 7, Zeile 2 ---	1-4,7,8
Y	DE 195 13 779 A (ZIMMERMANN & JANSEN GMBH) 17. Oktober 1996 (1996-10-17) Ansprüche 1-3 Abbildung 7 -----	1-4,7,8



Weitere Veröffentlichungen sind der Fortsetzung von Feld C zu entnehmen



Siehe Anhang Patentfamilie

\* Besondere Kategorien von angegebenen Veröffentlichungen :

"A" Veröffentlichung, die den allgemeinen Stand der Technik definiert, aber nicht als besonders bedeutsam anzusehen ist

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"L" Veröffentlichung, die geeignet ist, einen Prioritätsanspruch zweifelhaft erscheinen zu lassen, oder durch die das Veröffentlichungsdatum einer anderen im Recherchenbericht genannten Veröffentlichung belegt werden soll oder die aus einem anderen besonderen Grund angegeben ist (wie ausgeführt)

"O" Veröffentlichung, die sich auf eine mündliche Offenbarung, eine Benutzung, eine Ausstellung oder andere Maßnahmen bezieht

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"X" Veröffentlichung von besonderer Bedeutung; die beanspruchte Erfindung kann allein aufgrund dieser Veröffentlichung nicht als neu oder auf erforderlicher Tätigkeit beruhend betrachtet werden

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"&" Veröffentlichung, die Mitglied derselben Patentfamilie ist

Datum des Abschlusses der internationalen Recherche

Absendedatum des internationalen Recherchenberichts

10. Juli 2000

18/07/2000

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Bevollmächtigter Bediensteter

De Herdt, O

**INTERNATIONAL SEARCH REPORT**

Information on patent family members

International Application No

PCT/EP 00/02821

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